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On the production of element 102

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ABSTRACT

In 1957 the present authors reported the production of an isotope of element 102. By similar experiments made at Berkeley and in Moscow alpha disintegrations attributed to this element have also been found. At Berkeley, attempts to reproduce our results were unsuccessful. This has prompted us to make a thorough re-examination of our own experimental data.

The purpose of this paper is to comment on some comparisons between the Berkeley and the Stockholm experiments. Our re-investigation has not led to any new conclusions regarding the earlier interpretation of our results.

In 1957 we reported the production of an isotope of element 102 [1] in bombardments of curium targets with cyclotron-accelerated $^{13}C^{4+}$ ions. The reaction product which we ascribed to element 102 emitted alpha particles of about 8.5 MeV energy with a half-life of the order of 10 minutes. Although only about twenty decaying atoms of this product were detected, ion exchange experiments demonstrated that its chemical behaviour was consistent with predictions for element 102.

Since the publication of this work, two other groups have reported experiments on the production of element 102.

Ghiorso *et al.* [2] have bombarded curium with ^{12}C and ^{13}C ions accelerated in the Berkeley HILAC, but have not observed the element 102 isotope reported by us. By employing an improved recoil technique [3], they have, however, detected the isotope $^{254}102$ which has a half-life of 3 seconds. They identified it positively by establishing that the daughter product of its alpha decay is the known nuclide ^{250}Fm. In Moscow, Flerov *et al.* [4, 5, 10] have bombarded plutonium (^{241}Pu) with $^{16}O^{6+}$ ions, and have observed a short-lived product emitting long range alpha particles with an energy of 8.8 ± 0.5 MeV which they ascribe to an isotope of element 102. The alpha tracks were observed in nuclear emulsions, and no chemical identification was made.

The negative results obtained at Berkeley [2] appeared to cast some doubt on our results, and this has prompted us to make a thorough re-examination of our experimental data.

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The purpose of this paper is to comment on some comparisons between the Berkeley and the Stockholm experiments made in the paper of Ghiorso *et al.* [2].

We have not enough information to make an absolute comparison between the ^{13}C bombardments in the two different sets of experiments as regards for instance bombardment times and average currents used. From the ion current data given by Ghiorso *et al.* the maximum intensity of the ^{13}C beam used at Berkeley was about 2 or 3 times the maximum intensity used in Stockholm. However, neglecting a preliminary series, the number of ^{13}C bombardments of curium targets made in Stockholm was 62, each of about half an hour duration, in three series of experiments spread over a period of several months.

The Berkeley group reports a much higher yield of ^{246}Cf than can be accounted for only by their higher ion current, and they have taken this yield as a measure of the expected yield of element 102. However, they do not state whether the high yield quoted (40 counts per minute) was obtained with ^{12}C or ^{13}C ions. It is known that the cross section of the reaction which may be written $^{244}Cm(^{12}C, {}^{8}Be\,2n)^{246}Cf$ is many times higher than that of the reaction $^{244}Cm(^{13}C, {}^{8}Be\,3n)^{246}Cf$ for ions above 60 MeV energy [6].

In the Stockholm experiments reported, ^{13}C was used exclusively. Much higher yields of ^{246}Cf were obtained in ^{12}C bombardments which were not reported.

Apart from this, it is essential to consider the differences in target design. Products such as ^{246}Cf, probably produced mainly by direct interaction or stripping reactions, have rather short recoil ranges in the target material. The stacked foil target arrangement used at Berkeley in part of the experiments would be very efficient in collecting these recoils, since it consisted of several thin targets separated by catcher foils.

At Stockholm, a single, rather thick target was used, and a cover foil was interposed between the target and the catcher. It is probable, therefore, that a large proportion of the ^{246}Cf produced in our case was absorbed in the target or in the cover foil. Further, the ^{246}Cf yields obtained at Stockholm varied widely from one bombardment to another. For instance, in the experiment shown in Fig. 2 of our 1957 paper and discussed by Ghiorso *et al.*, the yield of ^{246}Cf was less than the maximum yield obtained by a factor of about 30. In general, high yields of ^{246}Cf were not obtained in those bombardments which gave the 8.5 MeV activity. This may be at least partly explained by assuming that the 8.5 MeV activity is produced by ions of lower energy than those which give a high cross section for the formation of ^{246}Cf. If this is so, the production of the 8.5 MeV activity by a $(^{13}C, 4n)$ reaction does not seem energetically unlikely.

It has been argued that our average carbon ion energy was too high to give appreciable yields of the $(^{13}C, 4n)$ reaction. We wish to emphasise that the energy figures given by us were very approximate. They were based on foil absorption measurements which indicated that in most bombardments the energy distribution had a peak in the range 80–100 MeV, but contained appreciable components also below 80 MeV. Cyclotron operating conditions could, however, affect the energy spectrum at a given radius, and it is possible that relatively high intensities of ions with energies below 80 MeV were present in at least part of the bombardments. The energy distribution in each individual bombardment was therefore not known accurately enough to enable definite conclusions to be drawn regarding the nuclear reactions producing the activity of interest.

Our curium targets had the following isotopic composition:

Mass number	Atom %
244	94.1
245	1.4
246	4.4
247	0.08
248	0.02

This is very close to the composition used at Berkeley in the experiments which yielded ^{254}102 from the (^{12}C, 4n) reaction on ^{246}Cm. It is therefore possible that ^{246}Cm (or with less probability one of the other heavy isotopes) may have been the vital target material at Stockholm also.

Regarding the ion exchange elution experiment shown in the same Fig. 2, the 8.5 MeV activity in drops 5, 6 and 7 was not eluted immediately after the free column volume as supposed by Ghiorso *et al.* [2]. The column was only 2 cm in height and 2 mm in bore, and the free column volume was less than 3 drops.

Calibration experiments using columns of the same dimensions and the same eluant solution have confirmed that mendelevium would have been expected in drops 8 and 9. The experiment therefore demonstrates that if the 8.5 MeV activity is due to an actinide element, it must be one of atomic number greater than 101. In a separate elution experiment, with hydrochloric acid as the eluant, it was shown that the 8.5 MeV activity eluted with the trivalent actinide elements present.

The 8.5 MeV activity was formed in very small amounts, but on analyzing the data, it was found to be quite significantly above the background in this energy range. Further the pulse analysis equipment was modified during the work, and the 8.5 MeV activity was observed with three different combinations of ionization chamber, amplifier, and pulse analyzer.

Several alternative explanations of the 8.5 MeV alpha activity have been considered, but none of these fits all the observed facts.

For instance, ^{225}Th could conceivably be formed by an unexpected type of stripping reaction, but if formed, such a product would have a very small recoil range, and it would not have penetrated the cover foil between the target and the catcher. Further, a thorium isotope would have been retained by the cation exchange column when the trivalent actinide were eluted with 6 N hydrochloric acid. Also, if the alpha activity of ^{213}Po, a daughter product of ^{225}Th, were present, it would not generally interfere with our 8.5 MeV activity because of the difference in alpha energies.

Other conceivable contaminants, such as ^{212}Po from ^{220}Em and ^{215}Em from ^{215}Fr have been considered, but all seem very unlikely for similar reasons.

The mass assignment made by us in 1957 was based on alpha decay systematics derived from data available at that time [7], and it appeared that a rather light isotope 251 or 253 was necessary to explain a decay energy as high as 8.6 MeV.

However, more recent information [8, 9, 10] seems to indicate that these predictions must be modified. The alpha decay energies of element 102 isotopes now appear to be higher than predicted. As a result, we feel that a decay energy of 8.6 MeV could correspond to a heavier isotope, for instance ^{255}102 which, as pointed out earlier in this paper, could have been formed by the (^{13}C, 4n) reaction in ^{246}Cm. This modification of the systematics could also have the effect of lengthening the predicted half-lives, and thus reducing the hindrance factor necessary to explain the observed half-life.

Thus our earlier mass assignment seems less certain, but apart from this, our re-examination of the experimental data has not led to any new conclusions regarding the interpretation of our results.

We feel that the differing results obtained in the above-mentioned laboratories can be explained only by further experimental studies including the properties of neighbouring nuclides. Until then we suggest that judgement on the discovery of element 102 should be reserved.

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